

Facile control of stretchability and electrical resistance of elastomer/polyaniline composites for stretchable conductors



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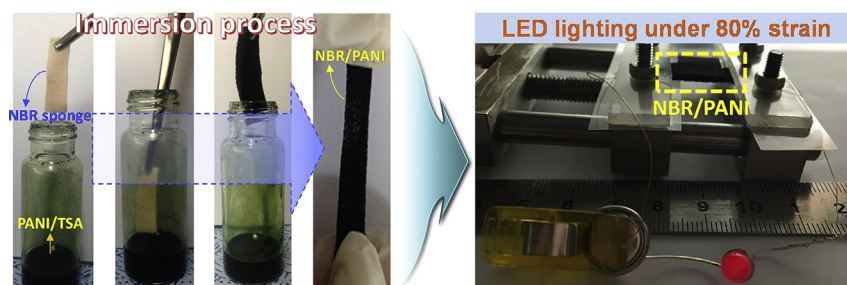
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HIGHLIGHTS

- Highly stretchable elastomer/PANI composites were fabricated using a facile immersion method.
- The weight fraction of PANI and the resistance of the composites could be easily controlled.
- NBR/PANI composites showed a high stretchability of 250% even after 7 immersion cycles.
- The composites exhibited a relatively small resistance change of 190% at a high strain of 210%.

GRAPHICAL ABSTRACT



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ABSTRACT

Porous elastomer/polyaniline (PANI) composites with high stretchability and adjustable resistance were successfully fabricated using a facile immersion method. Nitrile-butadiene rubber (NBR) and polyurethane (PU) sponges, which functioned as basic building blocks, were simply dipped into PANI solution. The weight fraction of PANI absorbed into the sponges monotonically increased with an increase in the number of immersion cycles, leading to the gradual decrease of electrical resistance. A NBR/PANI composite showed a high elongation at break of 250% even after 7 cycles of immersion and relatively small strain-dependent resistance change (190% at a 210% strain). This simple method to tune both mechanical stretchability and electrical resistance may be a useful means for the realization of organic stretchable conductors.

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1. Introduction

Stretchable conductors that would be replacements for traditional rigid conductors have attracted growing attention, due to

their potential application to diverse stretchable devices such as stretchable energy conversion/storage devices, biological sensors, electronic eye cameras, and stretchable light-emitting diodes [1–5]. Recently, three-dimensional (3D) conductive networks, which combine stretchable 3D network structures with conductive matters like conducting polymers and metal nanowires, have made a considerable progress. For example, Ge et al. have fabricated highly stretchable electronic conductors from polyurethane

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sponge-silver nanowire-poly(dimethylsiloxane) (PUS-AgNW-PDMS) composites by dipping PUS into AgNW solution and subsequently introducing PDMS into the PUS-AgNW sponge [6]. Chen et al. have also demonstrated high-performance supercapacitors composed of MnO₂ nanostructures-carbon nanotubes (CNTs)-sponge, using the similar “dipping and drying” method [7].

Polyaniline (PANI), which is one of typical conducting polymers, is widely employed as an active material for solar cells, gas sensors, and supercapacitors owing to the high conductivity, good availability of synthetic agents, and good environmental stability in atmosphere [8–12]. However, the poor mechanical stretchability of PANI limited its practical application to stretchable devices [13]. In recent years, many research groups performed intensive research to develop PANI-based conductive elastomers. For instance, Zhang et al. have fabricated highly stretchable buckled electrodes by attaching single-walled CNT (SWCNT)/PANI hybrid films to a pre-elongated PDMS substrate, which could be elongated as much as 140% [14]. Although these electrodes showed high stretchability and good repeatability, the fabrication procedure was rather complicated and the control of the relative weight of PANI to SWCNT was subtle. More recently, Dou et al. have rapidly synthesized a 3D nanostructured porous PANI hydrogel with tunable mechanical property, using amino trimethylene phosphonic acid as the gelator and dopant [15]. However, this PANI hydrogel was hardly elongated.

Herein, we report a facile method for fabricating elastomer/PANI composites with high stretchability and tunable resistance. Nitrile-butadiene rubber (NBR)/PANI composites showed particularly high stretchability of up to 250%. The simple immersion method employed in this study could easily control the amount of PANI absorbed in porous elastomers, thereby tune the mechanical property and the electrical resistance of the composites.

2. Experimental details

2.1. Materials

Aniline [C₆H₅NH₂] was purchased from Junsei Chemical Co., Ltd.; Ammonium persulfate (APS) [(NH₄)₂S₂O₈] was purchased from Kanto Chemical Co., Inc.; P-toluenesulphonic acid monohydrate (TSA) [C₇H₈O₃S·H₂O] was purchased from Sigma-Aldrich; Dimethyl sulfoxide (DMSO) [(CH₃)₂SO] was purchased from Wako Pure Chemical Industries, Ltd. These chemicals were used without further treatment. In this study, commercially available polyurethane (PU) and NBR sponges were used as basic frameworks for elastomer/PANI composites. PANI was synthesized using deionized water (DI water) as a solvent.

2.2. Synthesis of PANI/TSA

PANI/TSA was synthesized by a previously reported method [16,17]. 0.604 g of aniline and 1.235 g of TSA were dissolved in 10 ml of DI water. Then, 10 ml of APS (0.372 g) aqueous solution was added to the above solution as an oxidant. The mixed solution was shaken by hand for 30 s. After stabilized for 4 h at room temperature, the blackish green suspension was filtered and washed thoroughly with DI water, followed by drying at 60 °C for 12 h in vacuum oven.

2.3. Preparation of elastomer/PANI composites

Commercially available NBR sponge and PU sponge were cut into small pieces (~3.0 × 1.0 × 0.2 cm), and they were cleaned with DI water and ethanol several times. Then, they were dried at 80 °C for 2 h. Thus cleaned sponge pieces were immersed into the DMSO

solution containing a 2 wt% of PANI/TSA for 1 min. Then, they were taken out of the solution and dried at 80 °C for 2 h.

2.4. Characterization and measurement

Overall structures of NBR and PU sponges were first examined using an optical microscope (Eclipse 80i, Nikon). Microscopic structures of the initial sponges and their changes after immersion into the DMSO-PANI/TSA solution were analyzed using a field emission scanning electron microscope (FE-SEM, JEOL JSM-7500F). To check the absorption of PANI, Fourier transform infrared (FT-IR) spectroscopy (Bruker Vertex 70) was used. The resistance of elastomer/PANI composites was measured by a two-probe method under ambient condition. The stretching tests were performed to evaluate the stretchability of the composite samples using a home-made equipment. As a proof-of-concept test, a NBR/PANI strip was employed as a bridge connecting a light-emitting diode (LED) to a battery, and light emission from the LED was investigated depending on the strain applied to the composite strip.

3. Results and discussion

NBR and PU sponges are rubbery materials with unique porous structures. The smart combination of their rubbery nature and the porous structure could allow these materials to be substantially stretched. The microstructures of NBR and PU sponges and their deformations under an applied strain were observed using an optical microscope. As shown in Fig. 1, both sponges have well-developed frameworks with high porosity. The average pore size of NBR sponge is smaller than that of PU sponge, leading to the higher spatial pore density. On the other hand, the NBR framework shows rougher surface compared to the PU framework. When the two sponges are brought under tensile strains, the pores of the two sponges are elongated in the direction of the applied strain, while they are shrunk in the perpendicular direction (see the changes of the same pores indicated by red circles) [6,18]. These elongations are recovered to their initial states upon removing the applied strain. These stretching-recovery cycles were reversibly reproduced within maximum strains for the respective sponges.

The fabrication process of NBR/PANI and PU/PANI composites is simple enough, as shown in Fig. 2(a) and (b). It is composed of just two steps, soaking NBR and PU into PANI solution and drying the PANI-coated NBR or PU. During the soaking step, both sponges absorb PANI, thereby the original colors of the sponges turn into black resembling the color of PANI. At the same time, the volumes of the sponges are slightly expanded owing primarily to the swelling of DMSO. However, the swelling of DMSO turned out to weaken the mechanical strength of PU sponge, causing the sponge to significantly droop after taking it out of the PANI solution (see the third photo of Fig. 2(b)). This swelling effect is supposed to deteriorate the stretchability of the PU/PANI composite. The degree of swelling was relatively small for NBR sponge. As a consequence, the NBR piece shows a minimal length change after immersion into PANI solution followed by drying (compare Fig. 2(c) and (d) for relative length changes). Due to this difference in the swelling effect, NBR/PANI composites still retain high stretchability (see the length change of ~200% in Fig. 2(e) and (f)), whereas the stretchability of PU/PANI composites is considerably degraded (not shown). For comparison, pure PANI film was coated on a PDMS sheet (Fig. 2(g)). The PANI film is found to be easily cracked under stretching (Fig. 2(h)), proving its poor stretchability.

It is the PANI component to render the elastomer/PANI composites electrically conductive. In this regard, it would be important to examine the distribution of PANI inside the composites in relation to their frameworks. Fig. 3 shows SEM images of frameworks of

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